Report for 2003LA17B: Use of Synchrotron Microtomography and X-Ray Fluorescence to Better Understand Contaminant Diffusion in Reactive Barrier Systems

- Conference Proceedings:
 - Yin, M., C.S. Willson, and D.D. Reible, 2004, Investigation of Heavy Metal Migration from Contaminated Sediment to Capping material using XRF, presented at the 2004 AICHE Annual Meeting, November 7-12, 2004, Austin, TX.
- Other Publications:
 - Crannell, B.S., T.T. Eighmy, C.S. Willson, D.D. Reible, and M. Yin, 2004. Pilot-Scale Reactive Barrier Technologies for Containment of Metal-Contaminated Sediments and Dredged Materials. A Final Report Submitted to: The NOAA/UNH Cooperative Institute for Coastal and Estuarine Environmental Technology (CICEET), November 2004.
- Articles in Refereed Scientific Journals:
 - o Hartono, S. and C.S. Willson, 2004, Improvements in Groundwater Systems Analysis Through the Spatial and Temporal Estimation of Irrigation Pumping, submitted ASCE Journal of Water Resources Planning and Management.
 - Rahman, A., C.D. White, D. Carlson, and C.S. Willson, 2004, Improvements in Groundwater Flow Modeling Through the Integration of Resistivity Logs and Hydraulic Conductivity and the Use of Variogram Uncertainty. submitted Mathematical Geology.

Report Follows

Problem and Research Objectives

Sediment contamination can occur from the deposition of pollutants from the water column, deposition of contaminated particles, or through the seepage of contaminated groundwater. Unless removed or "controlled", contaminated sediments act as a continual source of pollutants to surface water bodies. This contamination may impact aquatic species and potentially render the water bodies unusable for recreation and/or drinking water supply.

Reactive barriers are capping materials that actively precipitate contaminants as they diffuse from contaminated sediment, rather than passively slowing the migration. The effectiveness of a reactive barrier depends on the ability to precipitate heavy metals from the pore solution onto the surface of the insoluble mineral by adsorption and surface precipitation. Continuous diffusion and subsequent precipitation within the reactive barrier also fills in the pore spaces around the materials, reducing permeability, and further inhibiting diffusion. The objective of this project is to continue the work where we are utilizing synchrotron X-rays to: (1) non-destructively quantify the diffusion of metals using X-ray fluorescence; and (2) quantify changes in the pore morphology of the reactive barrier using X-ray microtomography. Data and information obtained from these complementary experiments will help us better understand the pore-scale processes and phenomena impacting the transport of contaminant through reactive barrier systems. This work is also integrated with other research using X-ray absorption spectroscopy to look at metal speciation. The end result of this study will be better models and/or criteria for the design of reactive caps.

Methodology

X-ray Fluorescence (XRF): Basic XRF has become a well-established multi-element technique, capable of yielding accurate quantitative information on the elemental composition of a variety of materials. The technique is well-suited for studying environmental science problems because it is non-destructive, relatively rapid, and solids can be analyzed with little or no sample preparation. Apart from light elements, all elements with atomic numbers greater than 11 can be detected. The method is sensitive down to microgram-per-gram level and the results are precise and accurate if matrix effects can be corrected.

Synchrotron X-ray Microtomography (SXM): Synchrotron X-ray Microtomography has been developed over the past decade as a technique to non-destructively image the interiors of materials. Tomography deals with reconstruction of an object from its projections. Spatial resolutions on the order of ~ one to ten microns are possible because of the highly collimated and extremely bright X-rays produced by a synchrotron. These highly-parallel X-rays permit spatial resolution that is only limited by the optical components used to image them. Furthermore, the ability to tune to a monochromatic X-ray energy allows elemental discrimination.

In the spring of 2005, a multilayer monochromator was installed on the tomography beamline at CAMD. This monochromator will allow for high-resolution imaging of the sediment/reactive barrier cores.

Principal Findings and Significance

White-light and monochromatic XRF experiments have been performed at the LSU Center for Advanced Microstructures and Devices (CAMD) synchrotron facility. To date, these scans have been on cores obtained from a pilot-scale experiment currently being conducted at the University of New Hampshire. A final report to CICEET has been prepared (Crannell et al., 2004), one conference presentation has been made (Yin et al., 2004) and a manuscript is currently being prepared.

Metal sorption isotherm data is required to support the diffusion studies. Over the past year, a series of isotherm experiments at neutral pH values have been performed on sand, Anacostia sediment and two types of apatite for Cr(3), Cr(6), Zn, Cd, and Cu. These experiments were performed at neutral pH values. In addition, screening level diffusion tests were conducted to provide estimate of diffusion rates and concentrations.

Finally, based on the XRF data, we have been able to successfully fit a two-phase diffusion model and determine effective diffusivity values for a number of metals and sediment/barrier systems.